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Fuel processing activities at European level: A panoramic overview

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ARTICLE INFO

Article history:
Received 31 January 2014
Received in revised form
1 April 2014
Accepted 3 April 2014
Available online 3 May 2014

Keywords:
Hydrogen
Hydrocarbon fuels
Energy production
Fuel processor
APU
CHP

ABSTRACT

The present manuscript provides a panoramic overview of the most recent work carried out at a European level on the research and development of fuel processor (FP) units for various type of fuel cells (FCs), with an update on actual existing commercial products manufactured in Europe, namely auxiliary power units (APUs) and combined heat and power systems (CHPs). An increasing number of integrated complete FP units has been developed for a large variety of fuels: natural gas, biogas, low sulfur road diesel, propane, butane, liquefied petroleum gas (LPG), methanol, and ethanol. Some FP, APU and CHP systems are at present available on the European market.

Introduction

The expected future energy landscape will be mainly dependent on future energy demand, which is linked with the world's population growth and with the expanding energy requirements from industry and transportation sector. The world's population, in fact, is expected to increase from approximately 7.2 billion in 2012 to about 9.6 billion by 2050 (equal to a 33% increase) [1], with further long-term economic growth especially in non-OECD countries (Organization for Economic Co-operation and Development) [2].

World's energy demand will continue to grow in all sectors between now and 2040, as reported in Fig. 1. In particular, the highest energy demand growth rate is forecasted in the industry sector (mainly manufacturing, agriculture, construction, and mining), where the bulk chemicals and the petroleum refining industries consume most of energy. The transportation sector in non-OECD countries is expected to grow, too, but with a lower growth rate. An exception is forecasted for the OECD countries, where in the transportation sector the growth rate will be slightly negative, thanks to a significant decline in energy consumption by light duty vehicles (LDVs) and higher fuel efficiency compared to now [3]; more fuel-efficient new vehicles will, in fact, gradually replace older ones on the road. Within the transportation sector, only the energy demand for high duty vehicles (HDVs) is expected to increase, with the largest growth among all

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http://dx.doi.org/10.1016/j.ijhydene.2014.04.040

transportation modes (vehicles, aircrafts, marine vessels, railways) [3]. The residential sector, which accounts mainly for room heating and cooling, and water heating, will remain almost constant for OECD countries, whereas for the non-OECD countries it is expected to slightly grow. Finally the commercial sector is expected to slightly increase the energy consumed, for both OECD and non-OECD countries, in part for cooling and lighting commercial floor space, in part for installations of new data center servers for the information technology (IT) sector, which require high demand for ventilation and air conditioning [4,5].

According to the World Energy Outlook 2013 [2], renewable energy and nuclear power are the world's fastest-growing energy sources. However, fossil fuels and natural gas will continue to supply almost 80% of world energy through 2040, as evident from Fig. 2. Moreover, increasing supply of tight gas, shale gas, and coal-bed methane will support the forecasted growth for worldwide natural gas use [6,7]. It is expected that coal will grow faster than petroleum and other liquid fuels until 2030, mostly because of an increasing consumption of coal in China [8] and the high oil prices [2,9,10].

Energy consumption is an important issue of the global climate change debate [6,11–15]: recent monitoring of the CO_2 concentration in the atmosphere at Mauna Loa Observatory, Hawaii [16], has shown a constant increasing trend in the last years, as reported in Fig. 3, with a raise by about 2 ppm per year. Energy-related CO_2 emissions, produced by the combustion of liquid fuels, natural gas, and coal, account for much of the world's anthropogenic greenhouse gas (GHG) emissions [17].

Considering the expected energy growth trend, a secure and competitive energy supply will be a key challenge to meet our future energy requirements. Bearing in mind the environmental concerns linked with the emissions of GHGs, the most important actions to be taken into account are related to significantly slow-down the rate of energy-related CO_2 emissions [18–20], decouple CO_2 emissions from the economic growth [21–23], and favor a more innovative technological development, focused on highly fuel-efficient vehicles and industries [24,25].

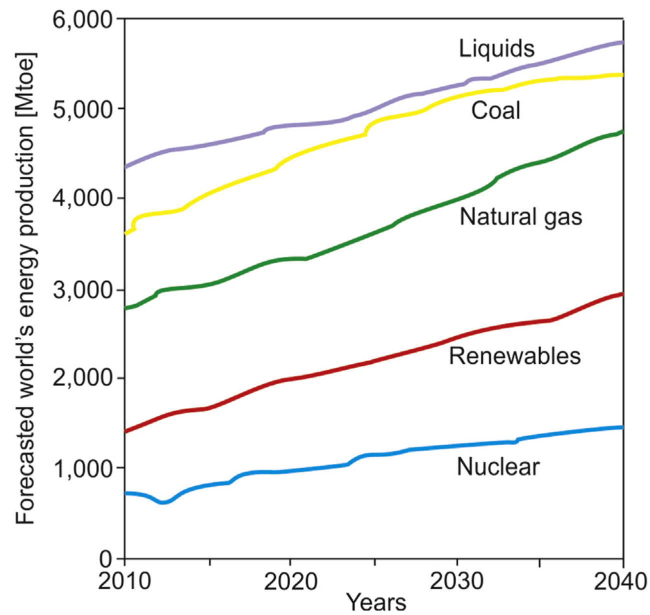

Fig. 2 – Forecasts of world energy consumption by fuel type in the period 2010–2040 [2].

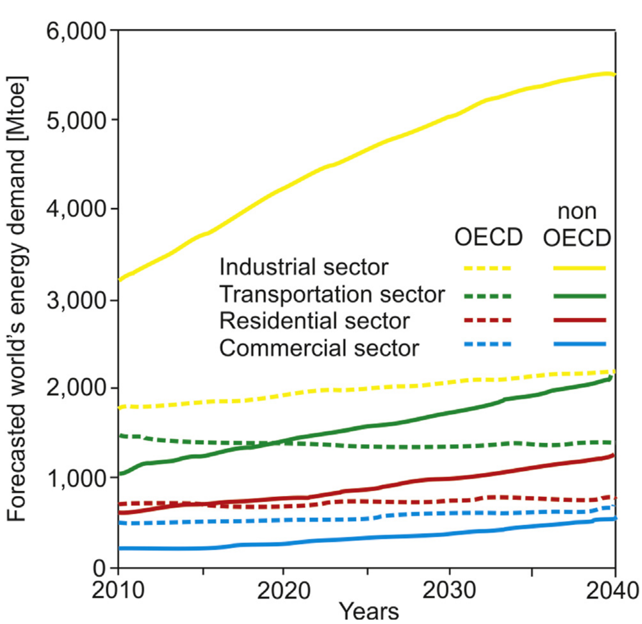

Fig. 1 – Forecasts of world energy demand by sector type in the period 2010–2040 in OECD and non-OECD countries [3].

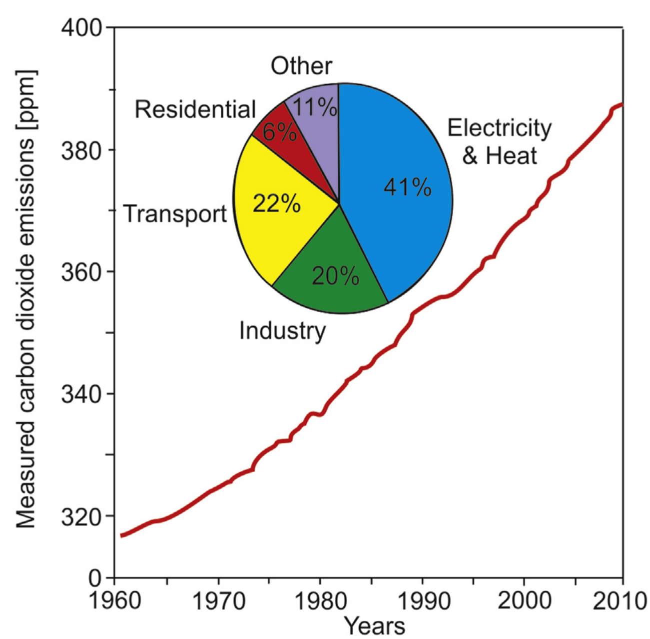

Fig. 3 – Mean CO_2 (ppm as a dry air mole fraction defined as the number of molecules of CO_2 divided by the number of all molecules in air, including CO_2 itself, after water vapor removal) at Mauna Loa Observatory, Hawaii [16]. In the onset: world CO_2 emissions by sector, in 2010 [17]. Other includes commercial/public services, agriculture/forestry/fishing activities, energy industries other than electricity and heat generation.

On such a context, the European Union's strategy for smart, sustainable and inclusive growth, the so-called "Europe 2020", aims to address safe energy supply, resource efficiency and climate change challenges by reducing GHGs emissions levels by 20% (or even 30% when possible) lower than 1990, increasing the share of renewable energies to 20%, and increasing the energy efficiency by 20% by 2020 [20,23,26,27]. One of the main instruments to achieve the aforementioned goals is the "Horizon 2020" program, the biggest EU Research and Innovation program over the period 2014 to 2020, which couples excellent science and industrial leadership, together with societal challenges [27].

The CO_2 emissions reduction target means the transition towards a low-carbon economy, which implies the almost complete decarbonization of Europe's power sector. The decarbonization process could be achieved along various pathways, such as nuclear energy, hydroelectric power, geothermal energy, solar energy, wind energy, the use of various kind of biomass, and the use of hydrogen as energy carrier to reach a true "hydrogen economy" [28–37]. The transition from current fossil-based to hydrogen economy includes two key elements: CO_2 capture and sequestration (CCS) with the utilization of solid carbon [13,38–43], and production of carbon-neutral synthetic fuels from bio-carbon and hydrogen generated from water using carbon-free sources, possibly employing renewable energies [37,44–58].

On this point of view, fuel cell (FC) technology and fuel processing of fossil and renewable fuels are playing a crucial role in future sustainable and distributed energy generation for mobile, portable and stationary applications. Fuel processing is the conversion of hydrocarbons, alcohol fuels and other alternative energy carriers to a gas product containing hydrogen [59,60]. Specifically, the employment of hydrogen on FC technology could ensure significant advantages in terms of efficiency and environmental impact, with reduced production of CO_2, representing thus an important alternative to the conventional energy production systems. Therefore, considering the actual lack of infrastructure for hydrogen production, storage and distribution, equipment operating with FCs fed with hydrogen produced by reforming of fossil fuel to generate power represent a valid and interesting alternative to overcome such an unfavorable situation [61–64]. Moreover, the reforming of fossil fuels could represent one practical option to create hydrogen filling stations realized with *on-site* fuel processor (FP) units fed with hydrocarbon fuels already present on the road, i.e. gasoline, diesel oil, natural gas and liquefied petroleum gas (LPG) [59,60,65–69]. This strategy would allow taking advantage of the existing fuel infrastructure with a limited environmental impact, and it would secure a smooth carbon–neutral transition from fossil-based to future hydrogen economy [24,34–37,70–73].

In this context, research and development on several reforming systems for FP units has gained a prevalent role in the perspective of solving these problems in a short to medium term. Moreover, FP is a viable option to meet the limited space demands on board of vehicles, specifically for auxiliary power units (APUs), when traction is not required, and to provide compact systems in stationary applications, precisely for combined heat and power units (CHPs) [59,69]. The present manuscript provides a panoramic overview of the most recent work carried out at European level on the research and development of FPs for various type of FCs, with an update on actual existing commercial products manufactured in Europe.

Fuel processing technology: R&D in Europe

A FP converts hydrocarbons, alcohol fuels or other alternative renewable fuels to a hydrogen-rich product gas, named reformate [59]. Depending on the reforming reactions, the reformate usually contains variable amounts of hydrogen, carbon monoxide, carbon dioxide, water, methane, and eventually nitrogen. A FP can be realized in a variety of configurations depending on its ultimate application. The carbon monoxide, in fact, can be partially or totally removed, depending on the final destination of the reformate gas [59]. Typically, the first processing step is accomplished by a reforming reactor, which can be a partial oxidation (POX) reactor, a steam reforming (SR) reactor, an autothermal (ATR) reformer, a thermal cracking (TC) reactor, a combination of POX and SR, called oxy-steam reforming (OSR) reactor, or a combination of POX, SR, and dry reforming (DR), namely a tri-reforming (TR) reactor, if the CO_2 is even a reagent [44,55,74–95]. The H_2/CO ratio in the reformate gas depends upon steam-to-carbon (S/C) and oxygen-to-carbon (O/C) molar ratios, which are determined by the fuel processed. Subsequent steps are targeted toward reformate conditioning, such as adjustment of the H_2/CO ratio and eventual CO removal. When high purity hydrogen is required, carbon monoxide has to be almost completely removed thorough a series of catalytic reactors for CO clean-up, consisting typically of a water gas shift (WGS) reactor and a preferential oxidation (PROX) or selective methanation (SMET) reactor [96–107]. The CO-PROX reaction removes carbon monoxide traces by adding air. However, this technology requires both a closely controlled low oxygen (or air) supply through mass flow meters, to keep at the lowest possible level the parallel hydrogen oxidation, and a wide operating temperature range for control purposes [69]. The CO-SMET reaction, instead, is able to reduce the carbon monoxide concentration below 10 ppmv without adding any co-reagent, by making the process inherently more easily controllable than CO-PROX. Moreover, the methanation reaction is less exothermic than carbon monoxide and hydrogen oxidations. However, the hydrogen consumption by the CO-SMET reaction is higher compared to the hydrogen consumed by the parallel oxidation inherently existing in the CO-PROX process [69]. Usually, an afterburner (AFB), or an integrated burner (IB), or a start-up burner (SUB) is present to burn the hydrogen exhaust gas from the FC anode and provide thus heat to the system. A series of balance of plants (BOP), such as heat exchangers for the internal heat recovery, water recovery condensers, air compressor, air cooler, water and fuel pumps, mixers, complete the whole FP [59,60,65,78,86,88].

Possible commercial applications of FPs can be envisaged in the realization of APUs and CHP units. APUs are realized by connecting together a FP and a FC, which can be a polymer electrolyte membrane (PEM) FC, a solid oxide (SO) FC, or a molten carbonate (MC) FC type, depending on the power output requested by the final application [108–137]. APU

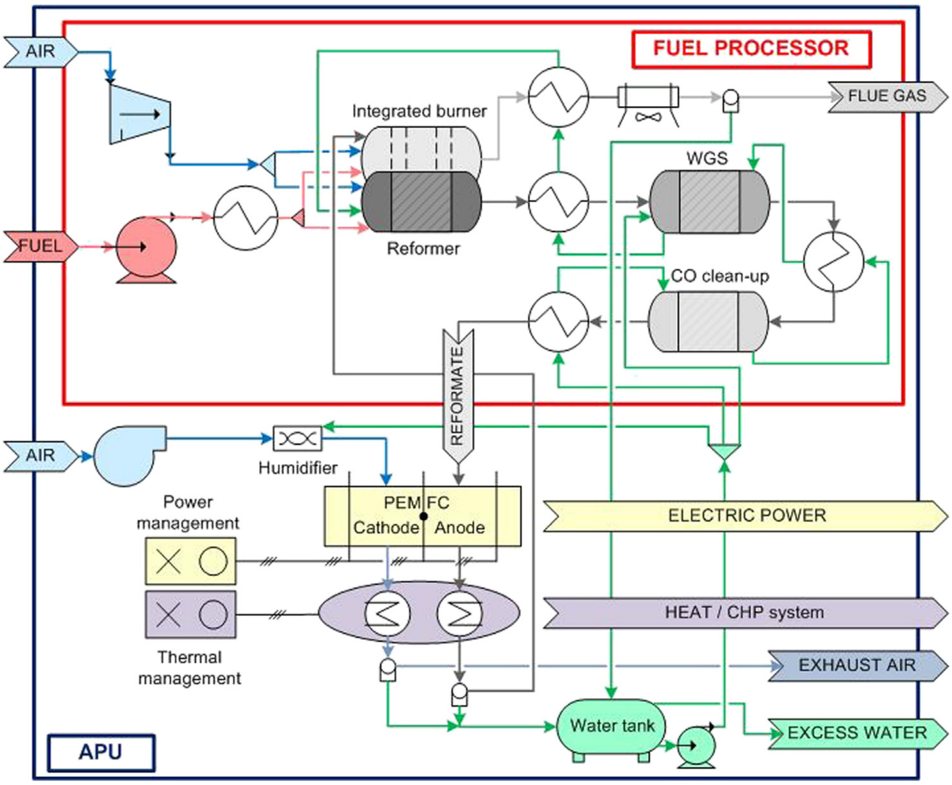

Fig. 4 – General scheme of a FP integrated into a PEM FC-based APU with CHP system.

devices can be placed onboard vehicles (cars/trucks, aircrafts, naval ships) to provide energy for functions other than propulsion: for recreational purposes onboard caravans and yachts, for trucks' idling during long stops overnight, for material handling vehicles as forklifts, or for running electrical systems onboard aircrafts. FC APUs offer much higher efficiency compared to conventional electrical power generators based on internal combustion engines (ICE) [59,117,137]. Aside from the benefit due to lower fuel consumption and CO_2 emissions, for applications onboard vehicles such systems can deliver electrical energy any time, even when the engine is switched-off, being independent from engine operation. In particular, truck APU's are a potential market for diesel-fed FC-based APU systems, considering the high demand for auxiliary power during trucks' idling overnight [115,117,127,129,130,134]. APUs can be even used as back-up and uninterruptible power systems (UPS) for mobile telecommunication industry and for military purposes [108–120].

CHP systems are cogeneration units based on APUs coupled with targeted heat exchangers and thermal power management units: while APU is producing electricity, the waste heat produced by the FC is simultaneously exploited to provide space heating, hot water (or steam) and, eventually, chilled water for air conditioning, for civil/commercial/industrial buildings and rural areas. As for APUs, CHPs cover a wide range of sizes and technologies [137–158]. The use of the cogeneration system as a distributed energy supply unit and thermal energy source has been widely studied for its high thermal efficiency: CHPs have proven to be successful in hospitals, hotels and private houses [140,148–150]. APUs and CHPs, in fact, are reported to create environmental, economic and social benefits, such as reduction in GHGs emission and operational cost, enhancing energy generation efficiency and energy supply security, and fostering a sense of responsibility on users [159–166].

A general scheme of a FP integrated within a PEM FC-based APU, along with the heat-exchangers required for the CHP system, is depicted in Fig. 4. Depending on the type of the reforming reaction (SR, ATR, OSR being the most used [84,85,90]), and on the type of FC present in the APU system (PEM, SO or MC FC [118,121,124,127,129,136,146]), the heat exchangers and the system design must be carefully chosen to optimize the heat recovery and improve the overall system efficiency. The FP and APU efficiency can be calculated according to the following equations [59,78,81]:

$$\eta_{\frac{FP}{gross}} = \frac{W_{CO} \cdot LHV_{CO} + W_{H_2} \cdot LHV_{H_2}}{W_{Fuel} \cdot LHV_{Fuel}} \tag{1}$$

$$\eta_{\frac{APU}{net}} = \frac{P_{FC}}{W_{Fuel} \cdot LHV_{Fuel} + P_{BOP}} \tag{2}$$

where W are the molar flows (of the CO and H_2 at the outlet of the FP, that is the valuable products of the reformate, and of the fuel entering the FP), and *LHV* the corresponding lower heating values, expressed in kJ mol^{-1}; P_{FC} is the net electrical power produced by the FC and P_{BOP} is the external power used for all of the mechanical items driven by electric motors, both expressed in kW. The net electrical power output of the FC, P_{FC}, can be calculated according to the following equation:

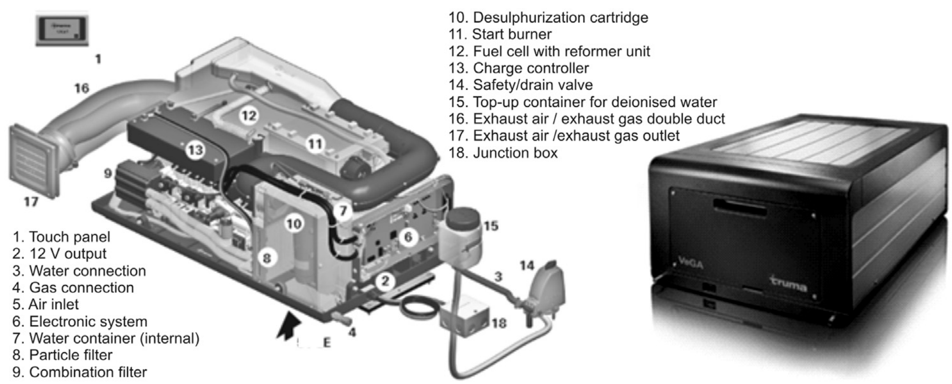

Fig. 5 – Commercial APU from Truma GmbH & Co. (Germany): scheme and picture of the VeGA fuel cell system [185,186].

$$P_{FC} = \eta_{anode} \cdot \eta_{FC} \cdot W_{H_2} \cdot LHV_{H_2} \quad (3)$$

with η_{anode} and η_{FC} the anodic efficiency and the FC efficiency, respectively [140,148,150].

The CHP efficiency is defined as the sum of the net electrical power output of the FC, P_{FC}, and the actual waste heat recovered, Q_{rec}, divided by the chemical energy input of the system:

$$\eta_{\frac{CHP}{net}} = \frac{P_{FC} + Q_{rec}}{W_{Fuel} \cdot LHV_{Fuel} + P_{BOP}} \quad (4)$$

For realizing FPs of small and medium size, ranging from a few produced kilowatts to a few hundred kilowatts, microstructured reactors (MSRs) appear to be very promising for achieving maximum compactness [55,59,60,65,66,78,83,88]. MSRs typically carry small channels, with dimensions in the sub-millimeter range, and a high surface-area-to-volume ratio, which reduces diffusive transport limitations. Thus, the technology offers many specific advantages as compared to conventional chemical reactors [59,60,65,78,88,95,167–170]: (i) enhanced heat transfer, which may be exploited for highly exothermic reactions with the aim of removing the heat generated and suppressing hot spot formation; (ii) superior mass transfer, which can help to optimally control gas concentration profiles in the reactor by suppressing the mass transfer influence on the overall kinetics as well as enhancing the gas adsorption and desorption phenomena at the heterogeneous catalyst surface; (iii) low pressure drop; and (iv) short residence times. When the reactor plates are coated with catalyst, the heat generated by exothermic reactions (or required by endothermic ones) may be removed (or supplied) by designing the reactor as a plate heat exchanger, thus improving the thermal management of the reactor itself [78,79,83,88,89]. Moreover, another way of exploiting the improved heat transfer is the combination of exothermic and endothermic reactions in a single reactor designed like a plate heat exchanger [170–181]. Thus, the process intensification benefits of micro-technology for gas-phase reactions are currently within the focus of the worldwide research related to fuel processing [59,60,65,78,81,88].

Germany, Truma GmbH & Co

Truma brand was founded in 1949, as manufacturing company for caravans' and motorcaravans' accessories as heaters and conditioning systems [185]. Actually, Truma Gerätetechnik company, located in Putzbrunn near München, is commercializing the "VeGA fuel cell system" [186–188], an APU system able to provide grid-independent electrical energy by the reforming of liquid camping gases. The system delivers a permanent power up to 250 W for all the necessary power requirements onboard caravans (lighting, conditioning, recreational activities, etc.). The system can be fed with 5, 11 or 33 kg cylinder LPG (propane plus butane). As shown in Fig. 5, the system includes a desulphurization cartridge for removing odorants (as the tert-butyl mercaptan, $C_4H_{10}S$) and other sulfur compounds (as the tetrahydrotiophene, C_4H_8S) present in the fuel for safety reasons in case of leakages; the start-up burner for heating up the fuel cell and reformer unit; the reformer unit for converting liquid gas into a hydrogen-rich gas; the fuel cell for electrochemical conversion of the hydrogen and air into electricity; the charge controller for charging the on-board battery of the recreational vehicle; a subsystem for supplying internal media (liquid gas, air, and water); the electronic control system for regulation and control; the supply air ducting for supplying reaction air and cooling air (with air filter); the system for removal of exhaust air and exhaust gas [179,185,186]. Available technical details are reported in Table 1 [186].

Greece, HELBIO S.A

HELBIO S.A. [189] is a spin-off company of the University of Patras, Greece. The company develops and sells hydrogen production systems, primarily from renewable sources, either for industrial use or integrated with low or high temperature (LT/HT) PEM FCs for APU systems and CHP production systems. The technology is based on steam reforming, followed by two stages of WGS reaction, a final CO clean-up based on CO-SMET reactor (residual CO concentration below 20 ppmv), and a pressure swing adsorption (PSA) system for the separation of hydrogen [189,190]. The multi-fuel systems are designed and manufactured for operation with both liquid and gaseous fuels (natural gas or liquefied petroleum gas), and bio-fuels (biogas and bioethanol). The delivered power output of the various products range from 1 to 20 kW for CHP systems, and from 300 W up to 5 kW for APU systems [189–192]. As for FP units producing hydrogen, the company supplies

Table 1 – Characteristic of the commercial products available on the European market.

Company	Technology	Fuel (fuel consumption)	Products	Technical characteristics	Dimensions ($L \times W \times H$ & weight)
Truma GmbH (Germany) [185]	Reformer + PEM FC	Propane (100 g h^{-1}) Butane (100 g h^{-1}) GPL (100 g h^{-1})	APU (VeGa fuel cell system) [186]	Max 250 W (6000 Wh day^{-1}), 20 A @ 12 V Operative temperature: −20 to +40 °C Operating gas pressure: 30 mbar Start-up time: 15 min	0.71 × 0.46 × 0.29 m^3; 40 kg (without water content)
HELBIO S.A. (Greece) [189]	Steam reformer + WGS + CO-SMET + PSA + LT/HT PEM FC	Natural gas (9.3–116 Nm^3 h^{-1}) Propane (6.9–86.5 kg h^{-1}) Biogas gas (15.7–195 Nm^3 h^{-1}) Ethanol (12–152 kg h^{-1})	FP [190]	FP-20: 20 Nm^3 h^{-1} FP-50: 50 Nm^3 h^{-1} FP-100: 100 Nm^3 h^{-1} FP-250: 250 Nm^3 h^{-1}	2 pieces, 5 × 2.4 × 2.5 m^3 3 pieces, 6 × 2.5 × 2.5 m^3 Custom design Custom design
			APU	300 W to 5 kW_e (up to 7 kW_{th})	5 kW_e: 0.55 × 0.35 × 0.4 m^3; 30 kg
			CHP	1–20 kW_e (25 kW_{th})	5 kW_e: 1.6 × 0.7 × 2.0 m^3; 320 kg
HyGear B.V. (The Netherlands) [196]	Steam reformer + WGS + PSA	Natural gas (26–30 Nm^3 h^{-1})	FP (HGS series) [197]	HGS-L: 42–25 Nm^3 h^{-1} Operative temperature: −10 to +35 °C Operative pressure: 8 bar Start-up time: max 30 min	4.05 × 2.43 × 2.59 m^3; 5500 kg
				HGS-C: 84–104 Nm^3 h^{-1} Operative temperature: −10 to +35 °C Operative pressure: 8 bar Start-up time: max 30 min	6.05 × 2.43 × 2.59 m^3; 9500 kg
SerEnergy A/S (Denmark) [199]	Steam reformer + HT PEM FC	Water/Methanol 40/60 vol.% (H3-350: 0.44 L h^{-1}) (H3-700: 0.88 L h^{-1}) (H3-5000: 1 L kW h^{-1})	APU (H3 series) [200]	H3-350: 350 W, 16.5 A @ 21 V_{DC} Operative temperature: −20 to +40 °C	H3-350: 0.27 × 0.20 × 0.59 m^3; 13.7 kg
				H3-700: 700 W, 24/48 V_{DC} Operative temperature: −40 to +50 °C	H3-700: 0.25 × 0.48 × 0.55 m^3; 27 kg
				H3-5000: 5 kW, 24/48/80 V_{DC} Operative temperature: −40 to +50 °C	H3-5000: 0.25 × 0.48 × 0.70 m^3; 45 kg
PowerCell AB (Sweden) [202]	ATR Reformer + PEM FC	Low sulfur road diesel	APU (PowerPac) [203]	Lab test (B1.0/2012): 2.6 kW_e, 24 V Operative temperature: +5 to +20 °C Start-up time: <60 min	400 dm^3; 230 kg
				Demonstration (B2.X/2014): 3 kW_e, 24 V Operative temperature: +5 to +35 °C Start-up time: <30 min	325 dm^3; 175 kg
				Truck product (2016): 3 kW_e, 24 V Operative temperature: −25 to +45 °C Start-up time: <10 min	250 dm^3; 150 kg
SOFCPower S.p.A. (Italy) [205]	CPOX reformer + SO FC	Natural gas LPG	CHP (EnGen®, HoTbox™) [206]	EnGen® 500: 0.5 kW_e, 8750 kWh Y^{-1}; 30–32% efficiency;	No data available
WS Reformer GmbH (Germany) [208]	FLOX® Steam reformers + WGS	Natural gas Biogas LPG Propane Methanol DME	FLOX®-Compact Reformers [209,211]	C1/FPMC1: 1 Nm^3 h^{-1} hydrogen delivered C4/FPMC4: 4 Nm^3 h^{-1} hydrogen delivered C10: 10 Nm^3 h^{-1} hydrogen delivered 78–81% efficiency Reformate quality: >75% H_2, <20 ppm CO Lifetime: >5000 h	C1: 290 × 360 × 480 mm^3, 30 kg C4: 400 × 400 × 800 mm^3, 40 kg C10: cylindrical shape 400 (diameter) × 1000 mm^2, (125 l), 90 kg
			FLOX®-Modular Reformers [209,211]	m50: 80 Nm^3 h^{-1} syngas delivered m100: 165 Nm^3 h^{-1} syngas delivered m200: 333 Nm^3 h^{-1} syngas delivered m400: 640 Nm^3 h^{-1} syngas delivered Syngas quality: 75% H_2, 13% CO, 8% CO_2, <4% CH_4, 300 °C, <15 bar	m50: 1400 × 1400 × 2500 mm^3, 2000 kg m100: 2000 × 1400 × 2500 mm^3, 2000 kg m200: 4500 × 1400 × 2500 mm^3, 7000 kg m400: 4000 × 1800 × 3500 mm^3, 10,000 kg

different systems capable to provide from 10 to 500 $Nm^3\ h^{-1}$, according to customer's needs. The hydrogen production technology is based on proprietary and patented catalytic reactor for reformation processes [193–195], which is based on the concept of the Heat-Integrated Wall-Reactor (HIWR), capable to offer very rapid heat exchange characteristics [89–91,190]. The HIWR concept scheme is reported in Fig. 6. The reactor consists of a bundle of tubes inside which reforming takes place on a catalyst deposited in a thin film on the inner surface of each tube. A combustion catalyst is deposited on the outer surface of the tube, to generate the heat of combustion very close to where the demand for heat is located, while heat transfer is very efficient across the metallic tube wall [190]. The designs incorporate very high degrees of heat integration enabling the reuse of all available heat either in the process (matching of cooling and heating streams) or as steam or hot water. Available technical details are reported in Table 1 [189,190].

The Netherlands, HyGear B.V

HyGear B.V. [196] is a Dutch company, located in Arnhem, established in 2002 and specialized in the downscaling of chemical processes, with special focus on miniaturized gas processing systems and on-site hydrogen generator systems (HGS) based on SR of natural gas, which can be installed at the end users' sites. The HGS generates hydrogen by reforming, using HyGear's proprietary reforming technology combined with highly efficient PSA units. Two models are available (HGS-L and HGS-C), with a nominal hydrogen output ranging from 42 to 104 $Nm^3\ h^{-1}$ [196–198]. The concept scheme and a picture of the commercial HGS-L unit is presented in Fig. 7. The systems are relatively big in size, they can work from an ambient temperature of −10 °C. Available technical details are reported in Table 1 [197].

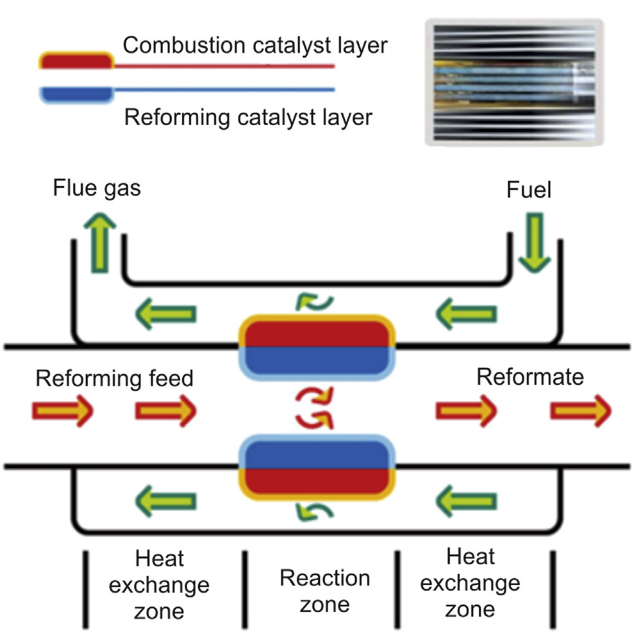

Fig. 6 – Reactor design concept of the commercial FP from Helbio S.A. (Greece): scheme and picture of the heat-integrated wall reactor (HIWAR) [189,190].

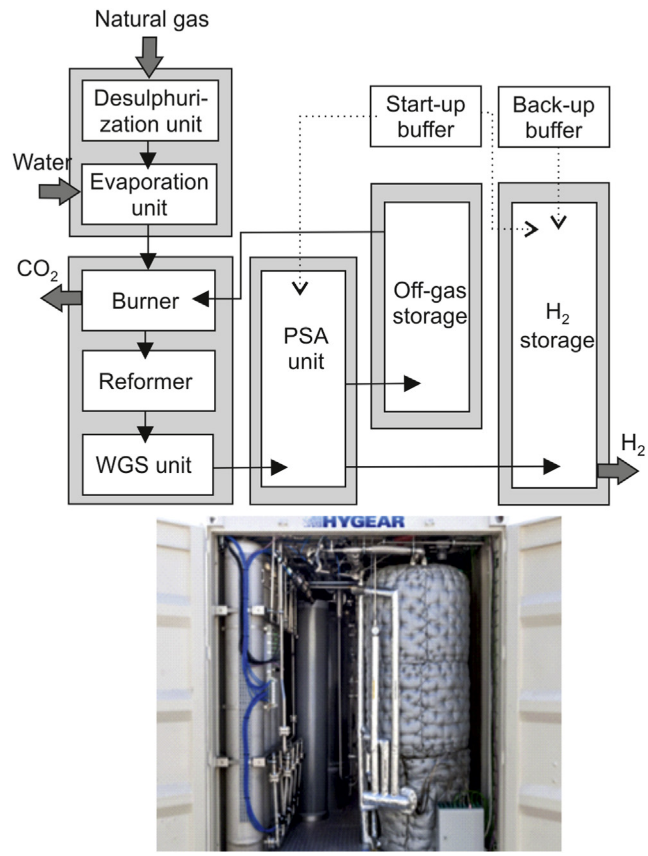

Fig. 7 – Scheme and picture of the commercial hydrogen generation HGS-L system from HyGear B.V. (The Netherlands) [196,197].

Denmark, SerEnergy A/S

SerEnergy A/S was founded in 2006 at the University of Aalborg, Denmark, as spin-off of the Institute of Energy Technology [199]. The company produces and commercializes HT PEM FCs and off-grid battery chargers. The latter consists of APU systems based on the steam reforming of methanol and a HT PEM FC. Methanol reforming typically takes place at 220–300 °C. Thanks to the use of an HT PEM FC there is no need of any additional gas clean-up, and the reformate can be directly fed to the stack. The off-grid battery charger are available in three systems (H3-350, H3-700, and H3-5000), able to deliver up to 0.35, 0.7 or 5 kW as electrical power output [200,201]. The APU reformates a solution of water/methanol 40/60 vol.%. A scheme of the commercial products is shown in Fig. 8. Available technical details are reported in Table 1 [200].

Sweden, PowerCell AB

PowerCell Sweden AB, located in Göteborg, was founded in 2008 as a spin-off of the Volvo Group [202], with the objective of bringing FC and fuel reformer technology to full commercialization. PowerCell's systems convert low sulfur road diesel to electricity in an energy efficient and environmentally friendly manner, resulting in reduced diesel consumption, lower emissions and silent operation. The main commercial

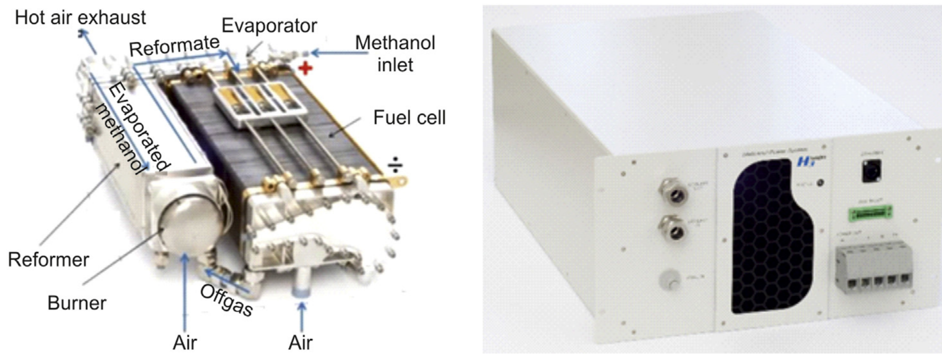

Fig. 8 – Scheme and picture of the commercial H3-5000 APU system from SerEnergy A/S (Denmark) [199,200].

items are the PEM FC stacks (S1 Fuel Cell, six models with a power output from 1 to 6 kW). The APU systems (PowerPac), based on a patented and proprietary system, are still under development [181,203,204]. The reforming technology is based on a catalytic autothermal reformer. The PowerPac is typically 20–30% more fuel-efficient than a diesel generator with batteries. A scheme of the PowerPac system is shown in Fig. 9. Available technical details are reported in Table 1 [203].

Fig. 9 – Scheme and picture of the commercial PowerPac APU system from PowerCell AB (Sweden) [202,203].

Italy, SOFCPower S.p.A

The company, situated in Mezzolombardo, was created in 2006 as spin-off company of the Eurocoating-Turbocoating Group, taking care of activities on SO FCs [205]. The company is currently focused on developing, producing and commercializing high efficiency proprietary SO FCs and CHP cogenerators for stationary applications in residential and commercial markets (EnGen® and HoTbox™ models [206,207]). Specifically, the EnGen® CHP systems are produced in two power ranges (up to 500 or 1000 W). The units operate with natural gas supplied by the gas network (methane or LPG). An electric heater assures the start-up of the system. Before entering the SO FC stack, the natural gas is pre-processed inside the system in a catalytic POX reactor. The SO FC cells, working at relatively low temperatures below 750 °C, are composed of thin-film YSZ (Yttria-Stabilized Zirconia) electrolytes sandwiched between two electrodes, a porous GDC/LSCF (Gadolinium Doped Ceria/Lanthanum Strontium Cobalt Ferrite) cathode and a Ni/YSZ anode [206]. At the SO FC stack outlet the exhaust gases are treated in a post-unit for combustion completion. Finally, the thermal energy of the flue gases is transferred to the water circuit in a heat exchanger, for domestic heating. An image of the EnGen® system is shown in Fig. 10. Available technical details are reported in Table 1 [206].

Germany, WS reformer GmbH

The company, situated in Renningen, close to Stuttgart, is a mid-size and independent company dedicated to innovative and highly efficient technologies based on the steam reforming of various fuels available in existing infrastructures (natural gas, biogas, LPG, propane, methanol, dimethyl-ether DME), born from the experience of the 20-years market company WS Wärmeprozesstechnik GmbH, pioneer and market leader in various segments of industrial high temperature processes [208]. WS Reformer GmbH realizes commercial reformers based on the FLOX®-Steam reforming process [209],

characterized by the patented FLOX®-heating process and the patented integrated reformate cooling system [210–215], plus the CO-shift unit, suitable for HT- and LT-PEM FC systems. A desulfurization unit upstream the reformer is also present [209,211]. In these reformers the evaporation of process water takes place by cooling down the reformate. With the patented FLOX® ("Flameless OXidation") combustion principle, the fuel gas and air are injected in such a way, that the reaction takes place downstream, evenly distributed in the combustion chamber. This leads to extremely low NO_x emissions and allows stable operation even with low calorific gases [210,214,215]. This results in a favorable thermal management (heating decoupling), which leads to simple control algorithms, high dynamics, simple and compact construction, high efficiency and low-emission operation [209]. The available commercial systems include a series of FLOX®-Compact Reformers, available in three different sizes (C1, C4 and C10 from 1 to 10 kW_e) plus an ultra-compact and ready-to-use Fuel Processing Module FPM-C1, as shown in Fig. 11A [211], suitable for micro-CHP in multi-family homes and business centers and for UPS power generators, and a series of FLOX®-Modular Reformers, suitable for on-site local hydrogen production, available in four different sizes (m50, m100, m200 and m400 from 20 to 400 $Nm^3\ h^{-1}$ hydrogen production), as shown in Fig. 11B [211]. Available technical data are reported in Table 1 [211].

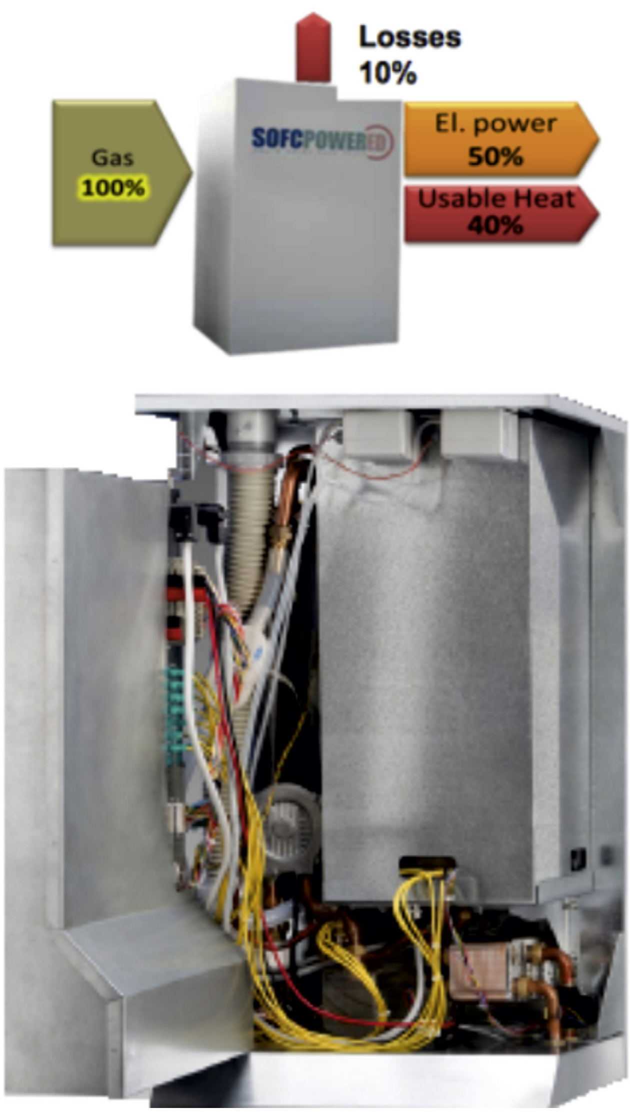

Fig. 10 – Scheme and picture of the commercial EnGen®500 CHP system from SOFCPower S.p.A.(Italy) [205,206].

Discussion

Sustainable and distributed production of energy and improving the utilization of fossil fuel resources have triggered R&D efforts in the recent decades. In this perspective, R&D on several reforming systems for FPs, APUs and CHPs has gained a prevalent role in the perspective of solving these problems in a short to medium term. The high efficiency coupled with the multiple technological options makes a FC-based APU the technology of choice for an engine-independent supply of electrical power in all kind of vehicles [59,64,111,115,117,137], or a FC-based CHP a viable solution to cogenerate heat and power in different environments [137–157]. Moreover, the application of micro-technology offers interesting technical solutions, allowing these systems to reach various commercial markets, depending on the final use, as shown in Table 1.

However, some practical problems still exist and must be carefully addressed to provide fully operating and smart technical solutions. The ongoing R&D has to address three major barriers: primarily the costs and, depending on the application, start-up and durability [59,64,65,111,117,170,180]. FPs and APUs require, in fact, high efficiency, stable operation and rapid start-up times for vehicle and portable applications.

As clearly evident even from the data reported in Table 1, APUs are able to deliver 250 W to 3 kW with a weight from 14 to 175 kg, depending on the manufacturer [186,190,200,203]. If micro-technology is not employed, as in the case of FPs realized by HyGear B.V. [196], the overall weight of a 100 $Nm^3\ h^{-1}$ hydrogen generator reaches 9500 kg [197]. It is interesting to note that the ongoing research already allowed weight and size reduction: considering the 3 kW APU prototype developed by PowerCell AB [202], the first lab test demo weighted 230 kg, while the actual 2014-prototype weights 175 kg, and a further reduction of weight is expected for the 2016-prototype [203].

Concerning the start-up procedure, usually it consists of four steps: igniting/preheating of the reforming reactor, heating the WGS reactor, initiating the CO clean-up process, and stabilizing CO emissions, before starting to feed the FC with hydrogen. Heating up a chain of reactors is a time- and energy-consuming process. Various start-up strategies are proposed in the literature: FP's items can be heated-up by burning a certain amount of the fuel in the burner usually present to recover the anodic exhaust. The hot exhausts are fed through the system heating up the chain of reactors of the FP, or on distributed parallel heat-exchangers, which is feasible when MSRs are adopted [59,60,65,66,86,88,91,181–184]. The start-up burner can be integrated with the reformer itself, or embedded into the reformer [113,122,124,167–170,209,211], to better exploit the developed heat of combustion, reducing thus the start-up time demand. For small-size APU systems, with nominal

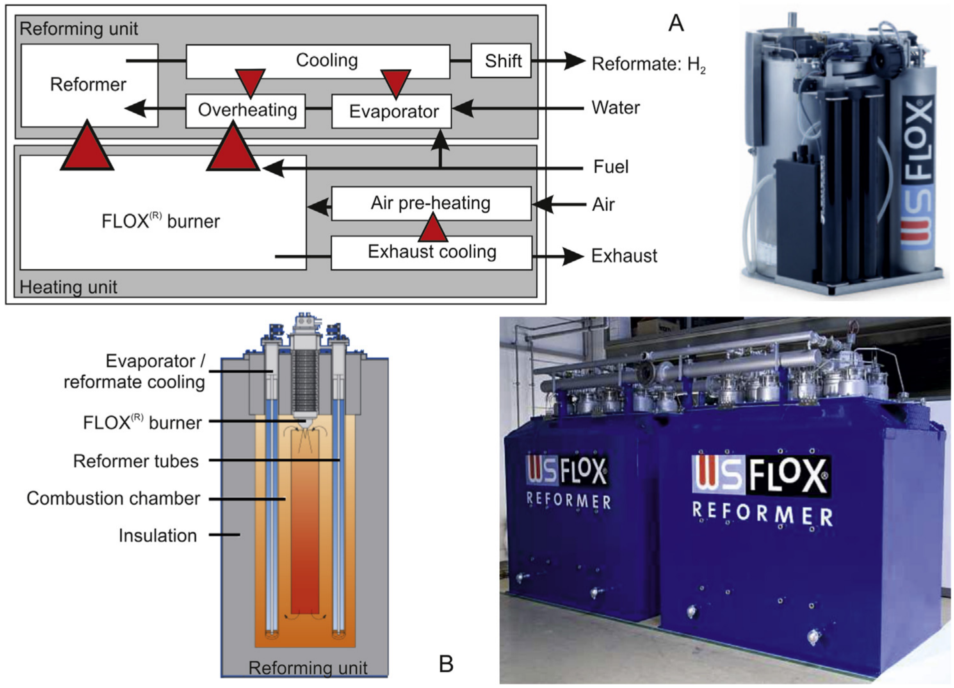

Fig. 11 – Schemes and pictures of the ultra-compact FLOX®-FPM C1 Steam Reformer (A: red triangles denote heat exchange areas) and FLOX®-Modular m200 (B) system from WS Reformer GmbH (Germany) [208,211]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

power output lower than 500 W, more sophisticated start-up procedures rely on *in situ* production of heat through the use of electric cartridges, connected to a battery pack [59,183,184]. At present, start-up times are still relatively long: one major limiting aspect of the system's dynamic behavior, in fact, is the inertia of reactors' thermal mass. This draw-back represents a great barrier for the application as automotive powertrain [124]. The amount of energy required to achieve full power in a FP can be expressed as a function of its thermal mass. Achieving shorter start-up time with a chain of reactors in series heated with hot gases is difficult. To decrease the time demand for start-up, the thermal mass of the components has to be as low as possible, and the start-up power has to be as high as possible. The lower the thermal mass of the FP, in fact, the lower the start-up time. The adoption of metallic monoliths or plate heat-exchangers create excellent heat transfer and consequently the start-up procedure results easier [59,95,96,172]. Thus, the concept previously mentioned of minimization of size and volume of a FP plays a favorable role even on the point of view of speeding-up the start-up time.

As appreciable from the data reported in Table 1, the available commercial items offer start-up time of approximately 30 min. It is worth noting that R&D efforts allowed reducing the start-up time demand by almost 50% (from Table 1, see the start-up time of the first lab test demo APU compared to the actual demonstrator by PowerCell [203]: from 60 to 30 min). In the open literature, as well, the reported start-up time of various FP or APU prototypes are relatively long, ranging from 60 to 30 min [111,115,117,118,124,129,148,150]. According to some simulation calculations, a potential reduction to 5–10 min appears feasible [111,124,180,181,203].

Conclusions and outlook

Considering the expected energy growth trend, and the environmental concerns linked with the emissions of greenhouse gases (GHGs), a secure and competitive energy supply will be a key challenge to meet our future energy requirements. Thus, the most important actions to be taken into account are related to significantly slow-down the rate of energy-related CO_2 emissions, decouple CO_2 emissions from the economic growth, and favor a more innovative technological development, focused on highly fuel-efficient vehicles and industries. Consequently, sustainable, distributed production of energy and renewable fuels and the improved utilization of fossil fuel resources have attracted increasing attention in the recent past. In this context, research and development on several reforming systems for fuel processor (FP) units has gained a prevalent role in the perspective of solving these problems on the short to medium term. FPs, in fact, represent a viable option to meet the limited space demands on board of vehicles, specifically for auxiliary power units (APUs), and to provide compact systems in stationary applications, precisely for combined heat and power units (CHPs). The application of micro-technology, in fact, offers suitable technical solutions. However, notwithstanding the first commercialization of FPs, APUs and CHPs, some practical problems are still existing and must be carefully addressed to provide fully operating and

smart technical solutions. The actual R&D has to address efforts to still overcome three major barriers: the costs, the start-up time and the durability.

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